

# Non-renewal counting processes in systems with ageing waiting times: microscopic origin of logarithmic time evolution

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We study a generic counting process for systems governed by ageing waiting times. In contrast to renewal continuous time random walks with independent waiting times, we consider the case when each state is characterized by its own internal waiting time process, having been initiated at time  $t = 0$ . Therefore each transition from state  $n$  to  $n + 1$  is triggered by  $\psi_1$ , the forward waiting time density, instead of the regular waiting time density  $\psi$ . For states characterized by heavy-tailed forms of  $\psi$  we obtain an asymptotically logarithmic evolution of the counting dynamics  $n(t)$ , whose fluctuations vanish relatively to the mean. The counting process introduced here describes the dynamics of crack propagation, the counting of on-off transitions in ensembles of blinking quantum dots, or, more generally, the dynamics in complex systems such as glasses or biological cells.

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In the classical Scher-Montroll-Weiss continuous time random walk (CTRW) model a new transition in the system, for instance, a nearest-neighbor jump on a lattice, occurs after a random waiting time [1, 2]. Individual waiting times are independent and drawn (renewed) from the common probability density  $\psi(\tau)$ . Power-law forms  $\psi(\tau) \simeq \tau^{-1-\alpha}$  with  $0 < \alpha < 1$  were originally introduced for charge transport in amorphous semiconductors [2]. In absence of a bias  $\psi(\tau)$  gives rise to subdiffusion  $\langle x^2(t) \rangle \simeq K_\alpha t^\alpha$ , where  $K_\alpha$  is the anomalous diffusion coefficient [3, 4]. Such long-tailed forms of  $\psi(\tau)$  were shown to govern the motion of tracer particles in the cytoplasm [5] and in membranes [6] of living cells, in reconstituted actin networks [7], as well as the blinking dynamics of single quantum dots [8] and the dynamics involved in laser cooling [11]. Physically, the form  $\psi(\tau)$  may arise from comb models [9] or random energy landscapes [10]. The divergence of the characteristic waiting time  $\int_0^\infty \tau \psi(\tau) d\tau$  leads to phenomena such as ageing [12] and weak ergodicity breaking [13], with profound consequences for our conceptions, e.g., in molecular cellular processes [14].

Here we relax the renewal property of CTRW processes and consider the counting process  $n(t)$  in a system in which waiting times are no longer a property of the walker in an annealed environment, but at the outset a property of each state  $n$ . The random quantity  $n$  could be the number of jumps in between independent CTRW processes in a completely biased random walk, see Fig. 1. At each  $n$ , events that can trigger a transition occur according to independent waiting time processes governed by  $\psi(\tau) \simeq \tau^{-1-\alpha}$  with arbitrary exponent  $\alpha$ , and we assume that all these processes were initiated globally at

time  $t = 0$ . As triggering events occur independently at different states, we typically arrive at  $n$  in between two triggering events. Arriving at  $n$  at some time  $t'$ , the time span  $\tau$  until the transition to  $n + 1$  is triggered is thus given by the forward waiting time density  $\psi_1(\tau|t')$ , which explicitly depends on the ageing time  $t'$  [12]. In the theory of renewal CTRW,  $\psi_1(\tau|t')$  governs the occurrence of the first jump in an aged system [12, 15, 16], while all successive waiting times are drawn independently from  $\psi(\tau)$ . We here consider a process in which *each* transition is controlled by  $\psi_1(\tau|t')$  and quantify the probability  $h_n(t)$ , and its associated moments, to find the system in state  $n$  at time  $t$ . Interestingly, we find a logarithmic growth of the moments with time and that the distribution  $h_n(t)$  becomes increasingly sharp for  $0 < \alpha < 1$  and  $\alpha > 2$ , while its width remains comparable to its mean in the intermediate regime  $1 < \alpha < 2$ .

As an example, consider the propagation of a linear crack front in a solid. At each point  $n$  measured in the propagation direction of the crack, a vacancy is moving diffusively perpendicular to the  $n$ -axis. If the crack has propagated up to site  $n - 1$  and the vacancy at  $n$  reaches the  $n$ -axis, the crack grows to  $n$ , etc. The local triggering process is thus given by a waiting time density of the form  $\psi(\tau)$  with exponent  $\alpha = 1/2$  corresponding to the return of a one-dimensional random walk [3]. At increasing  $n$ , and thus increasing time, the local triggering process on average ventures further away from the  $n$ -axis, such that waiting times typically increase with  $n$ : the process ages. In a complex system the triggering vacancy could itself diffuse with a power-law waiting time density with exponent  $0 < \gamma < 1$ , such that  $\alpha = \gamma/2$ . Alternatively, in an

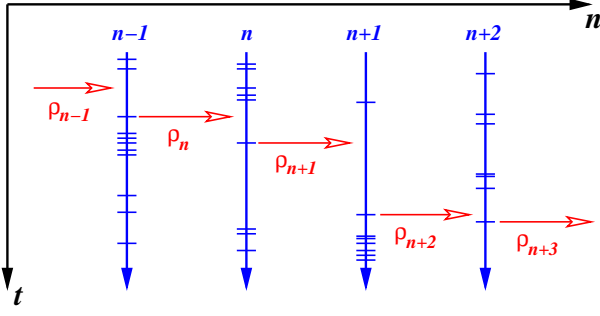


FIG. 1: Ageing counting process: propagation among successive states  $n$  as governed by the transition time density  $\rho_n(t)$ . At each  $n$  triggering events occur according to the waiting time density  $\psi(\tau)$ , globally initiated at  $t = 0$ . After the walker arrives to  $n$  at time  $t'$ , it has to wait for the next triggering event to be allowed to continue to state  $n + 1$  after the forward waiting time  $\tau$ .

ensemble of quantum dots or tracer particles in an actin network, we could be interested to count the number of events when the next quantum dot turns from an on to an off state or when the next tracer particle breaks out from its cage. As final examples we could consider the dynamics of train hopping in England, where the delay times indeed follow power-laws with exponent  $\alpha \approx 1.8$  [17], or message relaying along human communication paths. For instance, the Einstein and Darwin correspondences had delay times characterized by  $\alpha \approx 0.5$  [18]. A similar scenario could apply to cascades of active molecular transport in biological cells. The non-renewal counting process considered here is a general tool to quantify transitions in complex systems, in which such events depend on local triggering dynamics governed by power-law distributions of index  $\alpha > 0$ .

To formulate the model mathematically we introduce the probability density  $\rho_n(t)$  to arrive at state  $n$  at time  $t$ . This quantity fulfills the recursion relation

$$\rho_n(t) = \int_0^t \rho_{n-1}(t') \psi_1(t - t'|t') dt', \quad (1)$$

where  $\psi_1(\tau|t')$  is the density of the forward waiting time  $\tau$  that the walker has to spend in a new state after having arrived there at time  $t'$ . If the local triggering process follows  $\psi(\tau)$  with  $0 < \alpha < 1$  (see below for the case  $\alpha > 1$ ) then the waiting time  $\tau$  from time  $t'$  until transition to the next state is distributed according to the density

$$\psi_1(\tau|t') = \frac{\sin(\pi\alpha)}{\pi} \frac{t'^\alpha}{\tau^\alpha(t' + \tau)} \quad (2)$$

for the first jump in ageing CTRWs [15, 16, 19, 21]. If the walker is initiated in state  $n = 0$  at time  $t = t_0$ , the recursion (1) is closed by  $\rho_0(t) = \delta(t - t_0)$ . With respect to a short waiting time cut-off  $\tau_0$  in the power-law form of  $\psi(\tau)$ , Eq. (2) is the asymptotic result for  $t' \gg \tau_0$ .

Introducing the function  $\lambda(x) = \psi_1(x - 1|1)\theta(x - 1)$  with the Heaviside step function  $\theta(x)$ , Eq. (1) becomes

$$\rho_n(t) = \int_0^\infty \rho_{n-1}(t') \lambda(t/t') t'^{-1} dt'. \quad (3)$$

This expression can be interpreted as the transformation of the product of two independent random variables: denoting by  $\hat{t}_n$  the arrival time at state  $n$ , Eq. (3) states that  $\hat{t}_n = \hat{\chi}_n \hat{t}_{n-1}$ , where  $\hat{\chi}_n$  is independent of  $\hat{t}_{n-1}$  and has distribution  $\lambda(\chi)$ . In retrospect this is not surprising since  $t_{n-1}$  is the only time scale determining  $t_n$  [see Eq. (2)]. This means that we may write  $\hat{t}_n = \hat{\chi}_n \hat{t}_{n-1} = \hat{\chi}_n \cdots \hat{\chi}_1 t_0$ , or  $\ln(\hat{t}_n/t_0) = \sum_{i=1}^n \ln \hat{\chi}_i$ . Thus, in logarithmic time the transitions have independent waiting times. At large  $n$  we thus apply the central limit theorem. For  $\hat{z}_i = \ln \hat{\chi}_i$  we have the distribution

$$\langle \delta(z - \hat{z}_i) \rangle = e^z \lambda(e^z) = \frac{\sin(\pi\alpha)}{\pi} \frac{\theta(z)}{(e^z - 1)^\alpha}, \quad (4)$$

with the moment generating function  $G(p) = \langle e^{p\hat{z}_i} \rangle = \Gamma(\alpha - p)/[\Gamma(\alpha)\Gamma(1 - p)]$ . This defines the cumulants  $\kappa_m = (d/dp)^m \ln G(p)|_{p=0}$ . In particular, we find the average  $\mu \equiv \kappa_1 = -\Gamma'(\alpha)/\Gamma(\alpha) - \gamma$  and the variance  $\sigma^2 \equiv \kappa_2 = -\pi^2/6 + \partial^2 \ln \Gamma(\alpha)/\partial \alpha^2$ , where  $\gamma = -\Gamma'(1)$  is Euler's constant. For large  $n$  we then infer from the central limit theorem that  $\ln(\hat{t}_n/t_0)$  is normally distributed with average  $n\mu$  and variance  $n\sigma^2$ . Thus, for large  $n$  the approximate distribution assumes the Gaussian form

$$\rho_n(t) \sim \frac{1}{t\sqrt{2\pi\sigma^2 n}} \exp\left(-\frac{(\ln(t/t_0) - n\mu)^2}{2n\sigma^2}\right). \quad (5)$$

To proceed more systematically we apply a Mellin transform  $f(p) = \int_0^\infty t^{p-1} f(t) dt$  to Eq. (3). By the Mellin convolution theorem, Eq. (3) leads to  $\rho_n(p) = \lambda(p)\rho_{n-1}(p)$  in terms of the Mellin variable  $p$ , such that  $\lambda(p) = G(p - 1)$ . With  $\rho_0(p) = t_0^{p-1}$  we obtain  $\rho_n(p) = \lambda(p)^n t_0^{p-1}$ . While no simple expression for the inverse  $\rho_n(t)$  exists, it can be shown that the asymptotic behavior at  $t \rightarrow \infty$  is given by

$$\rho_n(t) \sim \frac{1}{(n-1)!} \left[ \frac{\sin(\alpha\pi)}{\pi} \right]^n \left[ \ln\left(\frac{t}{t_0}\right) \right]^{n-1} \frac{t_0^\alpha}{t^{1+\alpha}}. \quad (6)$$

Thus, the distribution of arrival times at state  $n$  mirrors the power-law tail of the original distribution  $\psi(\tau)$  plus a logarithmic correction with power  $n$ . Unsurprisingly the associated characteristic time scale diverges.

We are now ready to turn to the central quantity in this study, the probability distribution  $h_n(t)$  to find the system in state  $n$  at time  $t$ , equivalent to having arrived at  $n$  at  $t' < t$ , and not having moved since,

$$h_n(t) = \int_0^t \rho_n(t') \int_{t-t'}^\infty \psi_1(\tau|t') d\tau dt'. \quad (7)$$

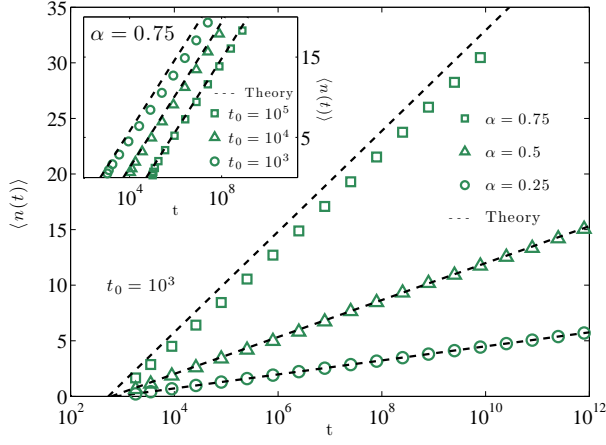


FIG. 2: Average counting process  $\langle n(t) \rangle$  versus time  $t$ . Symbols represent simulations for various  $\alpha$ , the dashed lines show the asymptotic behavior, Eq. (10) for  $q = 1$ . Simulations used  $t_0 = 10^3$  and  $\tau_0 = 1$ , with averages over  $10^7$  ensembles. Inset: convergence to the theoretical results with  $t_0$  for  $\alpha = 0.75$ .

The average state number  $\langle n(t) \rangle$  at time  $t$  and corresponding higher moments thus become  $\langle n^q(t) \rangle = \sum_{n=0}^{\infty} n^q h_n(t)$ . The Mellin transform of Eq. (7) is  $h_n(p) = \rho_n(p+1)[G(p)-1]/p$ , and therefore  $h_n(p) = t_0^p G(p)^n [G(p)-1]/p$ . To find the asymptotic behavior of the moments we note that  $G(p)$  is an increasing function of  $p$  (as long as  $p < \alpha$ ) which grows larger than unity when  $p$  is larger than zero. Thus the moments  $\langle n^q(p) \rangle$  diverge as  $p \rightarrow 0^-$ , and therefore have the fundamental strip  $-\infty < p < 0$ . The long time asymptotic behavior of the moments is therefore dominated by the singularity at  $p \sim 0$  [3]. Expanding  $G(p)$  at  $p \sim 0$  to second order,  $G(p) \sim 1 + \mu p + \frac{1}{2}(\sigma^2 + \mu^2)p^2$ . If we similarly expand the sum over  $n$  in  $\langle n^q(p) \rangle$  in deviations of  $G(p)$  from unity, making sure to keep the lowest and next to lowest order contributions, we find

$$\sum_{n=0}^{\infty} n^q G(p)^n \sim \frac{G(p)^q q!}{[1-G(p)]^{q+1}} + \frac{(q-1)G(p)^{q-1} q!}{2[1-G(p)]^q}. \quad (8)$$

Collecting terms we finally obtain the expression

$$\langle n^q(p) \rangle \sim \frac{\Gamma(q+1)t_0^p}{\mu^q(-p)^{q+1}} \left\{ 1 + \frac{p}{2} [\mu - q\sigma^2/\mu] \right\}, \quad p \rightarrow 0^-. \quad (9)$$

Inverting the Mellin transform, we find that at large  $t$

$$\langle n^q(t) \rangle \sim \left[ \frac{\ln(t/t_0)}{\mu} \right]^q \left\{ 1 + \frac{q}{2} \left[ \frac{q\sigma^2}{\mu} - \mu \right] \frac{1}{\ln(t/t_0)} \right\}. \quad (10)$$

The first two moments to leading order grow like  $\langle n(t) \rangle \sim \ln(t/t_0)/\mu$  and  $\langle n^2(t) \rangle \sim \ln^2(t/t_0)/\mu^2$ , respectively.

Fig. 2 compares the mean number  $\langle n(t) \rangle$  (10) with simulations, for the concrete form  $\psi(\tau) = \alpha\tau_0^\alpha/(\tau + \tau_0)^{1+\alpha}$ . Except for  $\alpha \rightarrow 1$  the simulations agree excellently with

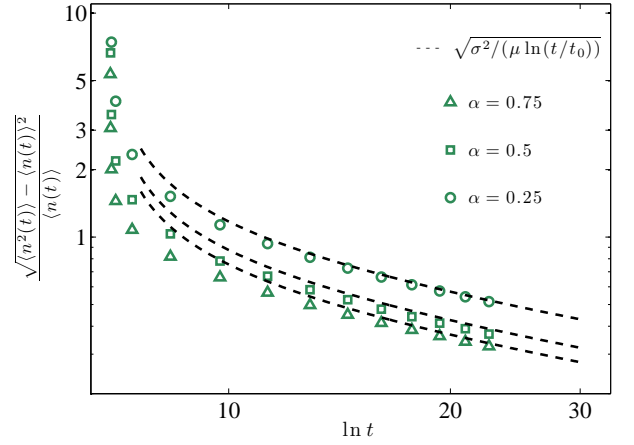


FIG. 3: Standard deviation versus mean as a function of time. Simulations (symbols) are compared to the asymptotic results for large  $t$  [Eq. (11), dashed lines]. Parameters used:  $t_0 = 10^3$ ,  $\tau_0 = 1$ , averaged over  $7 \times 10^5$  ensembles.

the asymptotic result (10). From the inset of Fig. 2 we see that the mismatch is due to the fact that  $t_0$  is not large enough, i.e., compared to  $t_0$  the distribution  $\psi_1(\tau|t_0)$  has not reached its asymptotic form, Eq. (2).

In Eq. (10) the  $q$ -dependence of the dominating term for the moments appears naturally, corresponding to a  $\delta$ -function behavior for the limiting distribution. From the subdominant terms of  $\langle n^q(t) \rangle$  we see that the deviation divided by the mean decays logarithmically as

$$\frac{\sqrt{\langle n^2(t) \rangle - \langle n(t) \rangle^2}}{\langle n(t) \rangle} \sim \sqrt{\frac{\sigma^2}{\mu \ln(t/t_0)}}, \quad (11)$$

as nicely corroborated from simulations in Fig. 3. This implies that the standard deviation of the counting process versus the mean number  $\langle n(t) \rangle$  becomes increasingly sharp, in contrast to the behavior of the position coordinate in biased subdiffusive CTRW processes, in which the ratio of deviation and mean tends to a constant [2].

To obtain the full form of the distribution  $h_n(t)$  we note that its Mellin transform  $h_n(p)$  can now be written as  $h_n(p) = t_0^p e^{n \ln G(p)} [G(p)-1]/p$ . In the large  $n$  limit,  $h_n$  is different from zero only for  $p \approx 0$ . To obtain an approximate result for  $h_n(t)$  we expand  $h_n(p)$  for large  $n$  and small  $p$ , keeping the product  $np^\nu$  constant, where the scaling exponent  $\nu$  is chosen as small as possible while still obtaining a non-trivial result when discarding the small terms. For  $\nu = 1$  a  $\delta$ -function is obtained for  $h_n(t)$ , for  $\nu = 2$  we find to zeroth order in small quantities  $h_n^{(0)}(p) = \mu t_0^p \exp[n(\mu p + \sigma^2 p^2/2)]$ , thus

$$h_n^{(0)}(t) = \frac{\mu}{\sqrt{2\pi\sigma^2 n}} \exp\left(-\frac{(\ln(t/t_0) - \mu n)^2}{2\sigma^2 n}\right). \quad (12)$$

This expression systematically improves by inclusion of higher order terms in  $p$  and  $np^3$ . To first order,  $h_n^{(1)}(p) =$

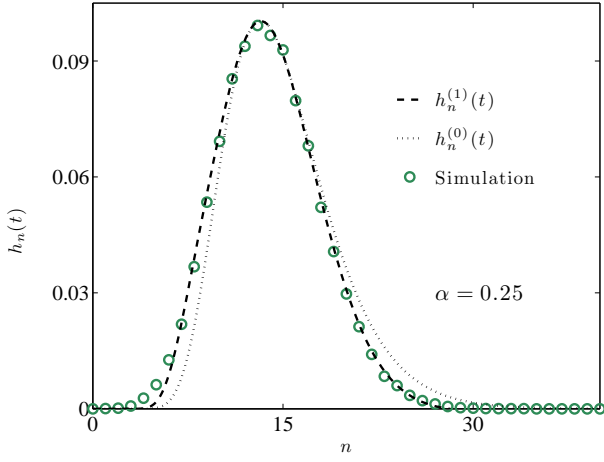


FIG. 4: Probability of being in state  $n$  at a given time  $t$ . Lines: analytical results from Eqs. (12) and (13), circles: results from simulations, averaged over  $7 \times 10^5$  ensembles. Parameters used:  $t = 7.9 \times 10^{12}$ ,  $t_0 = 10^3$ ,  $\tau_0 = 1$ , and  $\alpha = 0.25$ .

$h_n^{(0)}(p)[1 + (\sigma^2 + \mu^2)p/(2\mu) + \kappa_3 np^3/6]$ , thus

$$h_n^{(1)}(t) = h_n^{(0)}(t) \left[ 1 + \frac{\sigma^2 + \mu^2}{2\mu\sqrt{\sigma^2 n}} y + \frac{\kappa_3 n}{6(\sigma^2 n)^{3/2}} (y^3 - 3y) \right], \quad (13)$$

where  $y = [\ln(t/t_0) - \mu n]/\sqrt{\sigma^2 n}$ . In Fig. 4 we compare  $h_n^{(1)}(t)$  with simulations, demonstrating very good agreement for the dominating part of the distribution.

What is the behavior when  $\alpha > 1$ ? In this case  $\psi_1(\tau|t')$  has a finite limit  $\psi_1(\tau) = \lim_{t' \rightarrow \infty} \psi_1(\tau|t')$  given by  $\psi_1(\tau) = \langle \tau \rangle^{-1} \int_{\tau}^{\infty} \psi(\tau') d\tau'$  [21], where  $\langle \tau \rangle = \int_0^{\infty} \tau \psi(\tau) d\tau$ . Thus at long times one obtains  $\psi_1(\tau) \sim (\alpha - 1)A/[\langle \tau \rangle \tau^\alpha]$ . For  $1 < \alpha < 2$  this means that the counting process corresponds to the propagation as a one-sided Lévy flight on the time axis. Inverting this to find the distribution of the number of transitions to reach a given time will correspond to solving a first passage problem for one-sided Lévy flights. From this argument we find that the moments are given by [19, 22]

$$\langle n^q(t) \rangle \sim \frac{\Gamma(q+1)}{\Gamma(q(\alpha-1)+1)} \left( \frac{\langle \tau \rangle t^{\alpha-1}}{\Gamma(2-\alpha)A} \right)^q. \quad (14)$$

Thus for  $1 < \alpha < 2$  the fluctuations grow as fast as the mean  $n$ . For  $\alpha > 2$  we replace  $\alpha \rightarrow 2$  and  $\Gamma(2-\alpha)A \rightarrow \langle \tau^2 \rangle/2$ , such that  $\langle n^q(t) \rangle \sim (2\langle \tau \rangle t / \langle \tau^2 \rangle)^q$ . In this case the deviations vanish relatively to the mean. Thus, the deviations become increasingly negligible compared to the mean value  $\langle n(t) \rangle$  when the exponent  $\alpha$  is smaller than unity or larger than 2. In contrast their ratio saturates in the intermediate range  $1 < \alpha < 2$ .

The results we presented in Eqs. (7) to (13) apply more generally, beyond the ageing waiting time process considered so far. In fact, for Eq. (3) to hold we only require that the distribution  $\psi_1$  can be written in the form

$\psi_1(t-t'|t') = t'^{-1} \lambda(t/t') \theta(t-t')$ , which holds as long as there is no time scale in the problem other than the arrival time  $t'$ . Moreover, we require that  $G(p)$  can be Taylor expanded. An example when these two conditions are satisfied is the random walk model for transitions between energy minima in a simple glass proposed by Angelani et al. [23]. These authors found that the rate of transitions decays as  $c/t$ , where  $c > 0$  is a numerical constant. This decay rate of transitions corresponds to the waiting time distribution  $\psi_1(t-t'|t') = ct'^c \theta(t-t')/t^{1+c}$ , leading to the moment generating function  $G(p) = 1/(1-p/c)$ . Eqs. (7) to (13) therefore apply to this process with  $\mu = 1/c$ ,  $\sigma = 1/c^2$  and  $\kappa_3 = 2/c^3$ , and thus also in this case the fluctuations vanish relatively to the mean.

We finally consider the time average for a single realization  $n(t)$  of the counting process with  $\alpha < 1$ . Such time averages are routinely measured in single particle tracking assays [14]. For the number of states this time average is defined in terms of the lag time  $\Delta$  via [24]

$$\overline{n(\Delta)} = \frac{1}{t_2 - \Delta - t_1} \int_{t_1}^{t_2 - \Delta} [n(t + \Delta) - n(t)] dt, \quad (15)$$

where the observation time interval of the trajectory is from  $t_1$  to  $t_2$ . Averaging this behavior over many trajectories, the dominant behavior at  $t_1, t_2 \gg t_0$  becomes

$$\langle \overline{n(\Delta)} \rangle \sim \Delta \frac{1/\mu}{t_2 - t_1} \ln \frac{t_2}{t_1}. \quad (16)$$

for  $\Delta \ll t_2 - t_1$ . This linear behavior in  $\Delta$  contrasts the logarithmic time dependence of  $\langle n(t) \rangle$ . The linearity in  $\Delta$  and the dependence on the measurement time is similar to the findings of weak ergodicity breaking of renewal CTRWs with scale-free waiting time density [14, 25, 26].

We studied a general counting process in which successive transitions are affected by the age of the system through a local triggering process. This could be relevant for crack propagation in solids or counting statistics in ensembles of systems with power-law transitions times such as blinking quantum dots. We presented analytical and numerical results for the temporal distribution to reach a state  $n$  as well as the fractional order moments of the process  $n(t)$ . Interestingly the time evolution becomes logarithmic. Moreover, we found that the mean  $\langle n(t) \rangle$  of the counting process grows faster than the deviations of  $n(t)$  for power-law waiting time densities with exponent  $0 < \alpha < 1$  and  $\alpha > 2$ , while their ratio approaches a constant value in the intermediate case  $1 < \alpha < 2$ . We note that when the power-law exponent  $\alpha > 2$  the counting process becomes normal in the sense that the mean number grows proportionally to the process time,  $\langle n(t) \rangle \simeq t$ , and in this respect corresponds to waiting time densities  $\psi(\tau)$  with finite moments of any order, for instance, an exponential  $\psi(\tau) = \exp(-\tau/\tau_0)/\tau_0$ . However, similar to biased subdiffusive CTRW [27], the intermediate range  $1 < \alpha < 2$  still carries anomalous signatures.

Our results for the counting dynamics in an ageing system will be useful for the description of frustrated (e.g., glassy) systems, in which the renewal assumption of CTRW in an annealed environment no longer holds. As we showed, the generalized counting process exhibits weak ergodicity breaking. For the interpretation of single particle tracking experiments in such systems it will be interesting to obtain the full stochastic description of the position coordinate. We finally note that logarithmic system evolution was recently studied in the framework of *superheavy-tailed* waiting time densities within a renewal CTRW approach [28]. In contrast, our present results are based on a regular power-law waiting time density but with locally aged system update.

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